Efficient Simulation and Abuse Modeling of Mechanical-Electrochemical-Thermal Phenomena in Lithium-Ion Batteries

PI: Kandler Smith **Presenter**: Shriram Santhanagopalan

Team: Chao Zhang, Peter Graf, Ahmad Pesaran[*]
National Renewable Energy Laboratory
[*]**Now On detail to Vehicle Technologies Office**

Joshua Lamb
Sandia National Laboratories

Daniel Abraham, Pierre Yao, Dennis Dees
Argonne National Laboratory

Overview

This project was awarded in response to VTO FY15 Lab Call.

Timeline

- Project start date: Oct. 2015
- Project end date: Sept. 2018
- Percent complete: 60%

Budget

- Total project funding: $ 3.15M
  - DOE share: 100%
- Funding received in FY 2016:
  $1.05 M
- Expected Funding for FY 2017:
  $1.05 M

Barriers

- Gap between modeling tools and cell design process in the industry
- Lack of simulation tools integrating mechanical failure and abuse response of batteries for practical assessment of battery safety
- Limited understanding of complex failure mechanisms resulting in expensive over-design of batteries

Partners

- Argonne National Laboratory (ANL)
  - Pouch Cells and data for parameter estimation
- Sandia National Laboratories (SNL)
  - Cell-level mechanical abuse testing for validation of mechanical models
- Forming Simulation Technologies, Ohio State University, George Mason University
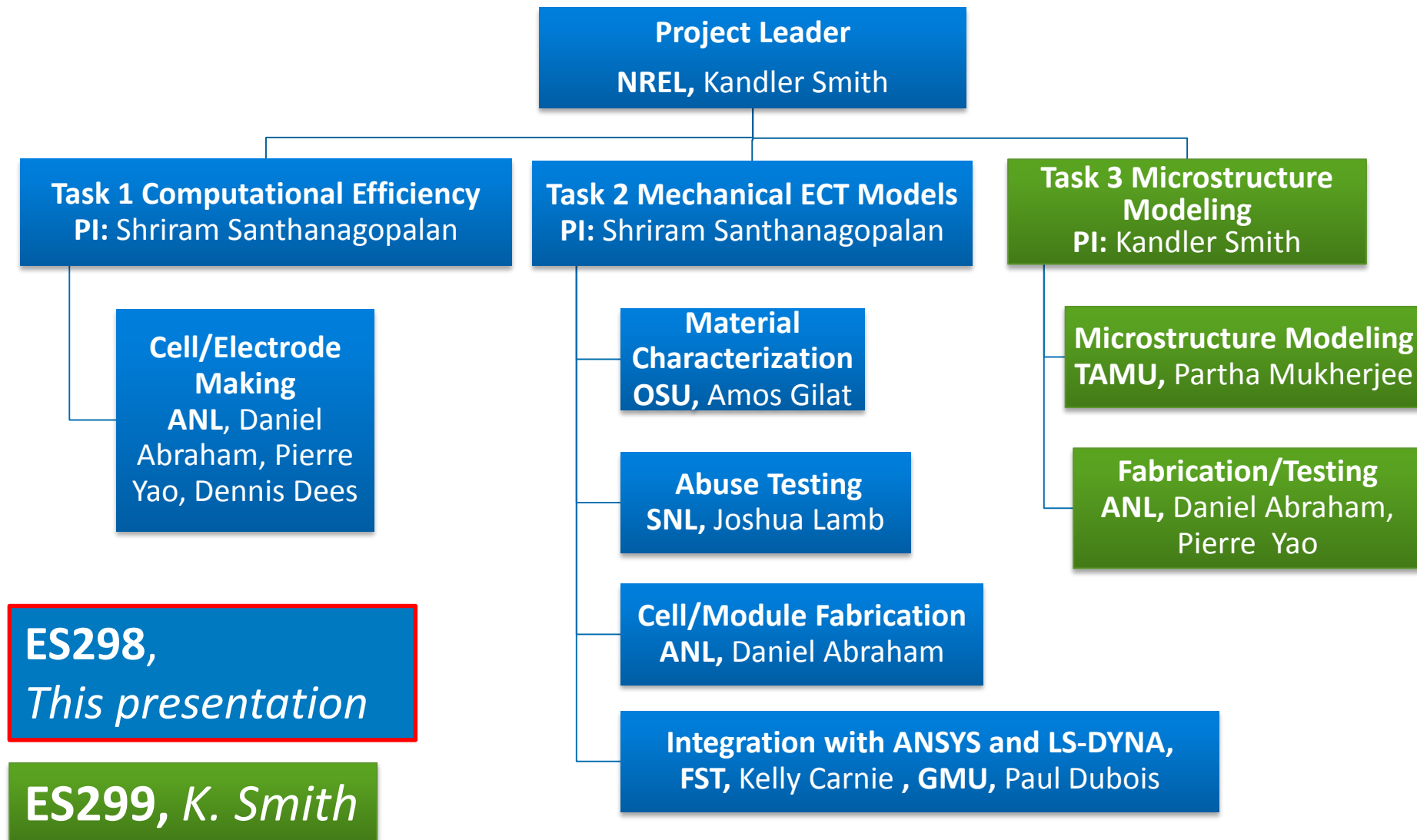 - Integration with ANSYS and LS-DYNA

Background and Motivation

- VTO launched the Computer-Aided Engineering of Batteries (CAEBAT) project to develop validated modeling tools to accelerate development of batteries, in support of vehicle electrification R&D to reduce dependence on imported oil.

- Over 40 different end users from the community have adapted the Multi-Scale Multi-Domain (MSMD) modeling approach developed under CAEBAT.

- Feedback from the first few sets of end-users has helped us identify priorities that will enable wider use of model-based design:

  o Standardize <u>identification of the model parameters</u>

  o Increase <u>computational efficiency</u>

  o Extend the models to include <u>mechanical failure</u> of cells and packaging components

  o Close gaps between <u>materials R&D</u> and CAEBAT modeling tools
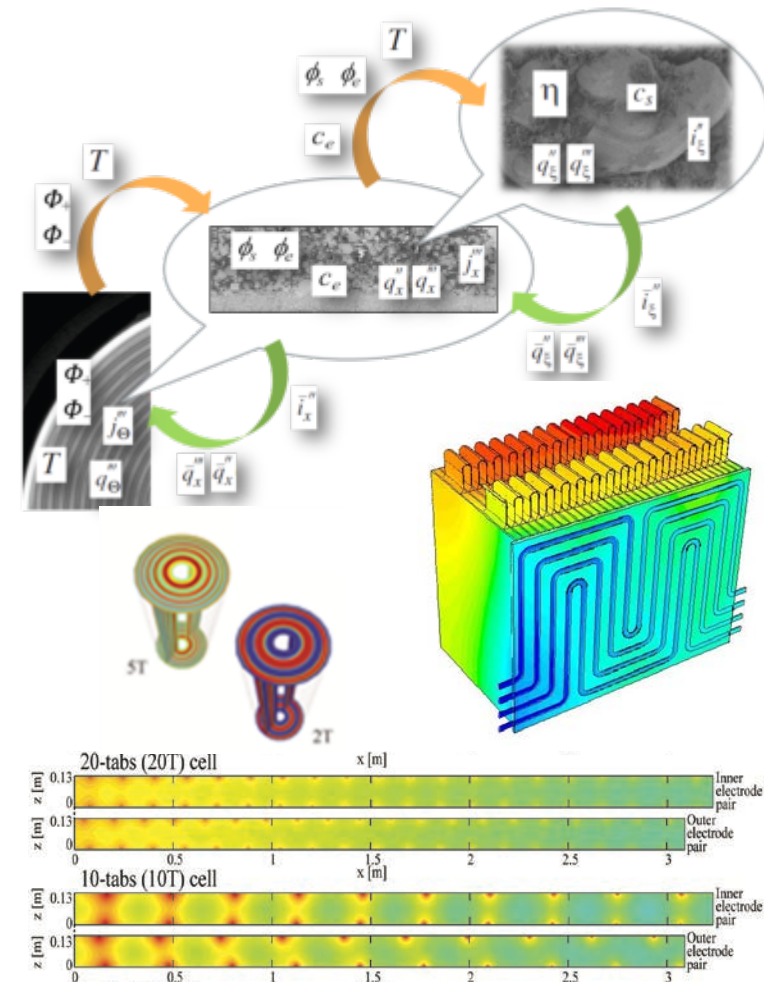
MSMD models previously developed in CAEBAT have been widely adapted in the community and helped us identify gaps.

Project Structure

Project Title: Computer-Aided Battery Engineering Consortium

Project Leader
NREL, Kandler Smith

Task 1 Computational Efficiency
PI: Shriram Santhanagopalan

Task 2 Mechanical ECT Models
PI: Shriram Santhanagopalan

Task 3 Microstructure Modeling
PI: Kandler Smith

Cell/Electrode Making
ANL, Daniel Abraham, Pierre Yao, Dennis Dees

Material Characterization
OSU, Amos Gilat

Microstructure Modeling
TAMU, Partha Mukherjee

Abuse Testing
SNL, Joshua Lamb

Fabrication/Testing
ANL, Daniel Abraham, Pierre Yao

Cell/Module Fabrication
ANL, Daniel Abraham

ES298,
This presentation

ES299, *K. Smith*

Integration with ANSYS and LS-DYNA,
FST, Kelly Carnie **, GMU,** Paul Dubois

Relevance

## Objectives for March 2016 – March 2017

Computational Efficiency:

- Demonstrate 1000-fold increase in computational speed using model order reduction methods
- Document efficiency enhancement approach for deployment on to commercial software platforms

Parameter Identification:

- Develop and document the procedure to extract parameters for the MSMD models

Mechanical Models:

- Present initial demonstration of simultaneous coupling between mechanical failure and the thermal response of the cell during a crush test

Initial demonstration of efficient thermal, electrochemical, and mechanical models
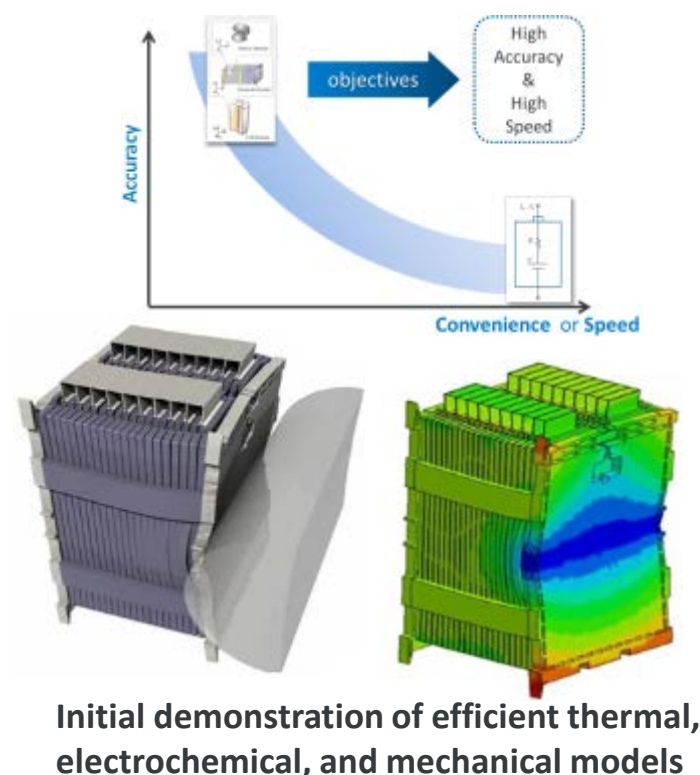
Impact: By making disruptive CAE design tools available on desktop computers for use by the battery community, this effort supports the following goals identified by the VTO:

1. Expedite path to $ 125/kWh electric vehicle (EV) battery costs by drastically reducing the number and duration of battery design cycles in the industry
2. Reduce module/pack costs by maximizing insight gathered on failure modes in batteries, from a limited subset of tests currently performed

Milestones

	Milestone Name/Description	Deadline	Milestone Type	Status
Comp. Effic.	M 1.1 Draft summary documentation of GH-MSMD framework	8/31/2016	Qrt. Prog. Meas.	Done
	M 1.2 Validate GH-MSMD using half cell data from ANL	1/31/2017	Qrt. Prog. Meas.	Done
	M 1.3 Present at the DOE Annual Merit Review	6/30/2016	Qrt. Prog. Meas.	Done
	M 1.4 Perform out design evaluation and performance evolution study using newly developed multiphysics GH-constituent models	7/31/2018	Qrt. Prog. Meas.	On track
Mech. Abuse	M 2.1 Demonstrate simultaneous coupling in MECT model that shows interaction of mechanical deformation with the thermal response of the cell under different strain-rates within 10% error against data	3/31/2016	Annual SMART (Go/No-Go)	**Go**
	M 2.2 Detailed documentation describing the mechanical tests procedure for development and validation of constitutive models for individual battery components and battery cells with < 5% error on the mechanical response at the component level between data and models	7/31/2017	Annual SMART (Go/No-Go)	On track
	M 2.3 Interim update on mechanical models demonstrating damage propagation across multiple axes of battery cells and battery modules	12/31/2017	Qrt. Prog. Meas.	On track
	M 2.4 Report summarizing model validation for MECT simulations	4/30/2018	Qrt. Prog. Meas.	On track

Task 1 - Computational Efficiency

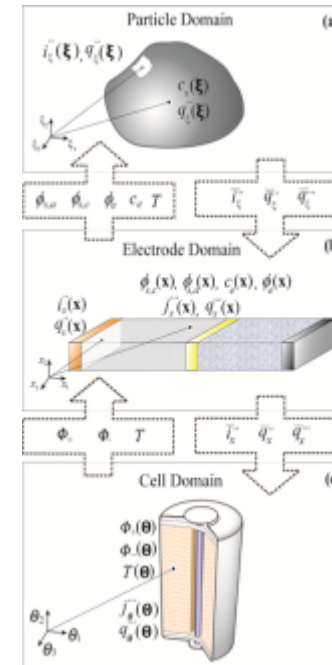Step 1: Nonlinear Multiscale Implicit Formulation

$$\phi = f(i; \boldsymbol{x}, \boldsymbol{p})$$

Step 2: Timescale Separation & Variable Decomposition

$$\phi = g(i; \boldsymbol{x}, \boldsymbol{p}) + h(i; \boldsymbol{x}, \boldsymbol{p})$$

Step 3: Partial/Selective Linearization

$$G(i; \boldsymbol{x}, \boldsymbol{p}) = \frac{dg}{di}$$

$$\phi = G(i; \boldsymbol{x}, \boldsymbol{p})i + H(i; \boldsymbol{x}, \boldsymbol{p})$$
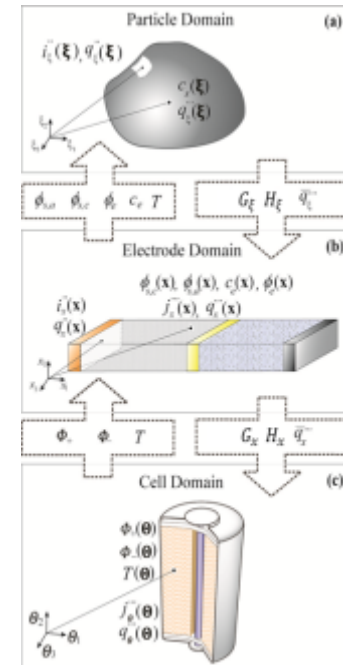
MSMD (Previous work)   **GH-MSMD (New)**

Efficient and Extensible Quasi-Explicit Modular Nonlinear Multiscale Battery Model: GH-MSMD

Gi-Heon Kim, Kandler Smith,[*,z] Jake Lawrence-Simon, and Chuanbo Yang[*]

National Renewable Energy Laboratory, Golden, Colorado 80401, USA

Complex physics and long computation time hinder the adoption of computer aided engineering models in the design of large-format battery cells and systems. A modular, efficient battery simulation model—the multiscale multidomain (MSMD) model—was previously introduced to aid the scale-up of Li-ion material & electrode designs to complete cell and pack designs, capturing electrochemical interplay with 3-D electronic current pathways and thermal response. This paper enhances the computational efficiency of the MSMD model using a separation of time-scales principle to decompose model field variables. The decomposition

G.-H. Kim et al., *J. Electrochem. Soc.*, A1076-88 (2017)

The selective G-H linearization approach drastically reduces computational burden!
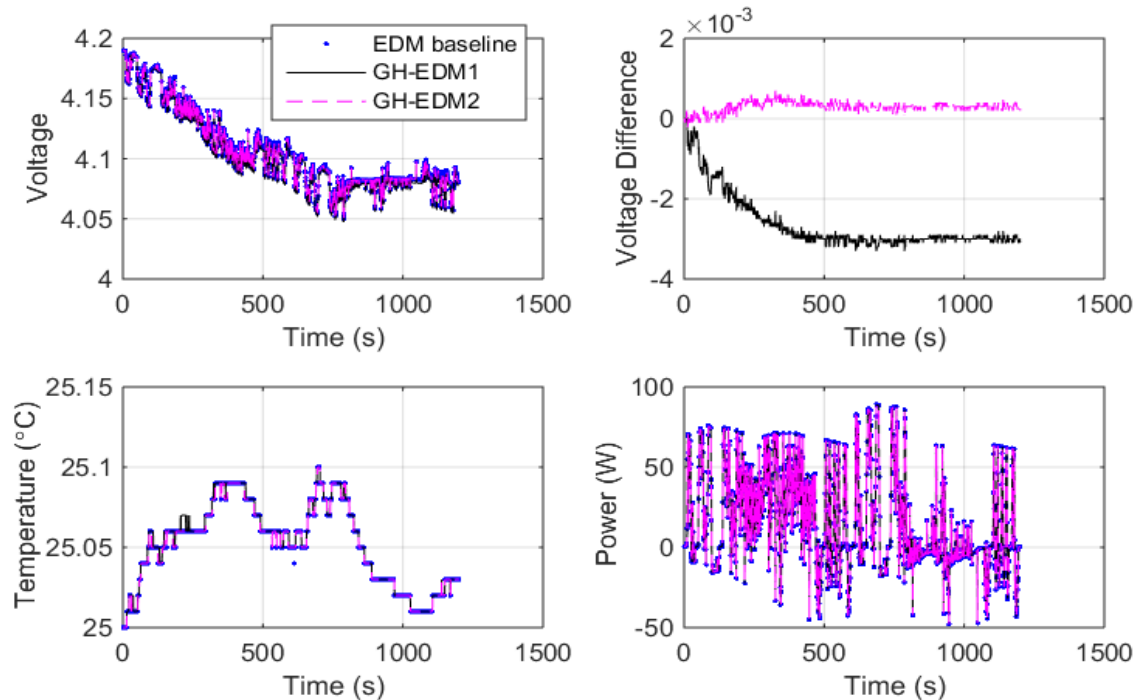
Simulation case		Computation time for Electrode Domain Models (EDM) in seconds		
Load Profile	Temperature (°C)	EDM baseline	GH-EDM1	GH-EDM2
1C	25	360.13	3.03	0.44
1C	0	816.21	3.50	0.47
Drive cycle	25	1205.92	7.06	0.83
Drive cycle	0	8786.45	45.00	1.27

Model Parameter Identification Workflow

Pre-processing and filtering of raw data

Experimental set up to cycle cells for collecting data

Format data from native formats for battery cyclers

Setup baseline MSMD Inputs

Fitting of model to data

$$\frac{\partial}{\partial x}\left(\kappa^{eff}\frac{\partial}{\partial x}\right) + \frac{\partial}{\partial x}\left(\kappa_D^{eff}\frac{\partial}{\partial x}\ln c_e\right) + j_x''' = 0$$

$$\kappa^{eff} = \kappa\varepsilon_e^p$$

$$\kappa_D^{eff} = \frac{2RT\kappa^{eff}}{F}(t_+^0 - 1)\left(1 + \frac{d\ln f_\pm}{d\ln c_e}\right)$$

$$\frac{\partial(\varepsilon_e c_e)}{\partial t} = \frac{\partial}{\partial x}\left(D_e^{eff}\frac{dc_e}{dx}\right) + \frac{1 - t_+^0}{F}j_x''' - \frac{i_e^*}{F}\frac{\partial t_+^0}{\partial x}$$

MSMD-Model

$C_{Li}{}^{max} = 4.905037e+04 \pm 7.067772e+01\ mol/m^3$
$D_s{}^{ref} = 4.392799e-15 \pm 2.562774e-17\ m^2/s$
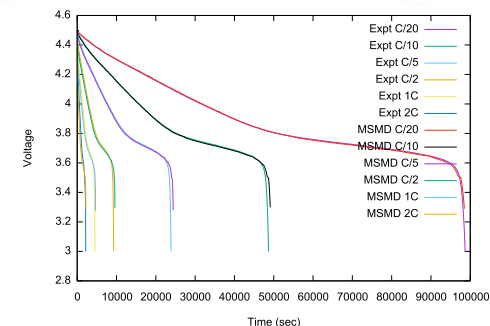
Calibrated Model and Parameters

- Python script parses data to meet model needs
- Parameter estimation based on Levenberg-Marquardt algorithm
- Workflow independent of model(s)/data set(s)
- Can use the same approach for multiple models and/or datasets – as long as the list of inputs and outputs are standardized (e.g., using the OAS)
- Process can be easily wrapped with a GUI as workflow stabilizes

Material Data and Cells from CAMP Facility at ANL

Material specifications for the cell components provided by ANL include:
- Electrode composition
- Thickness, porosity, loading density
- Particle size and distribution
- Current collector thicknesses

Figure Credit: Dan Abraham, ANL

1 µm
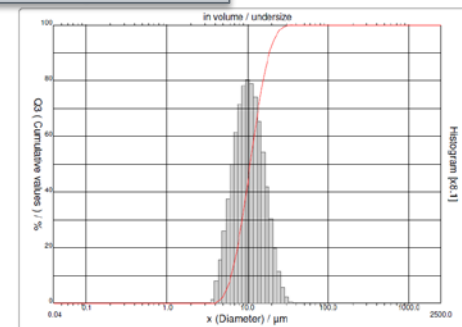
~15 µm sized **secondary** particles contain many **primary** particles

A-C015(+) is matched to A-A002A(-) for 4.4 V full cell cycling

A-C015(+): made by CAMP (NCM523)	A-A002A(-): made by CAMP
Positive Electrode:	**Negative Electrode:**
90 wt% $Li_{1.03}(Ni_{0.5}Co_{0.2}Mn_{0.3})_{0.97}O_2$	91.8 %wt ConocoPhillips: CGP-A12 graphite
5 wt% C45 (Timcal)	2 wt% C45 (Timcal) + 0.17 %wt Oxalic Acid
5 wt% PVdF binder (Solvay 5130)	6%wt KF-9300 Kureha PVDF binder
9.17 mg/cm^2 loading density - coating	5.88 mg/cm^2 loading density - coating
8.25 mg/cm^2 loading density - active/oxide	5.51 mg/cm^2 A12 graphite loading density
33.5% electrode porosity	38.4% electrode porosity
34-µm-thick composite coating	44-µm-thick composite coating
20-µm-thick Al current collector	10-µm-thick Cu current collector

CFF-B24A-P5

Baseline electrolyte: 1.2M $LiPF_6$ in EC:EMC (3:7, w/w)
Baseline separator: Celgard 2325 (trilayer, PP/PE/PP)

Pouch cells with 300 mAh nameplate capacity
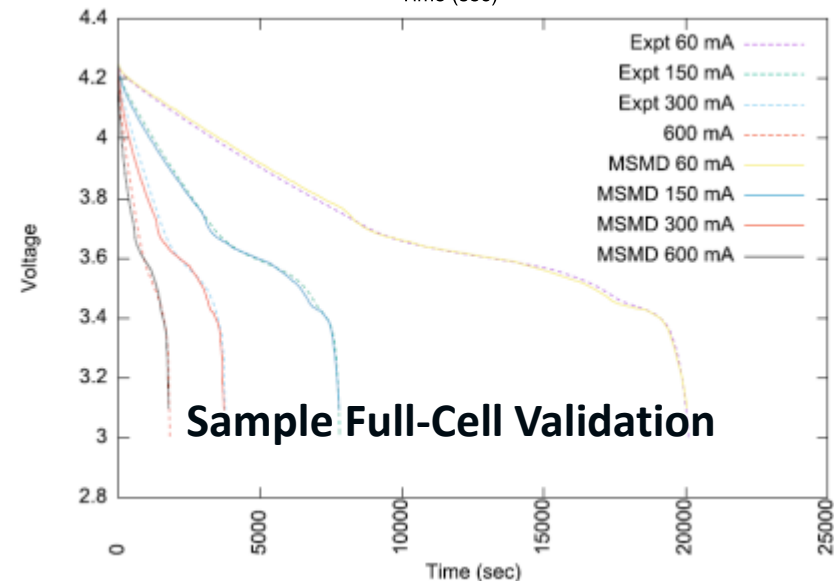
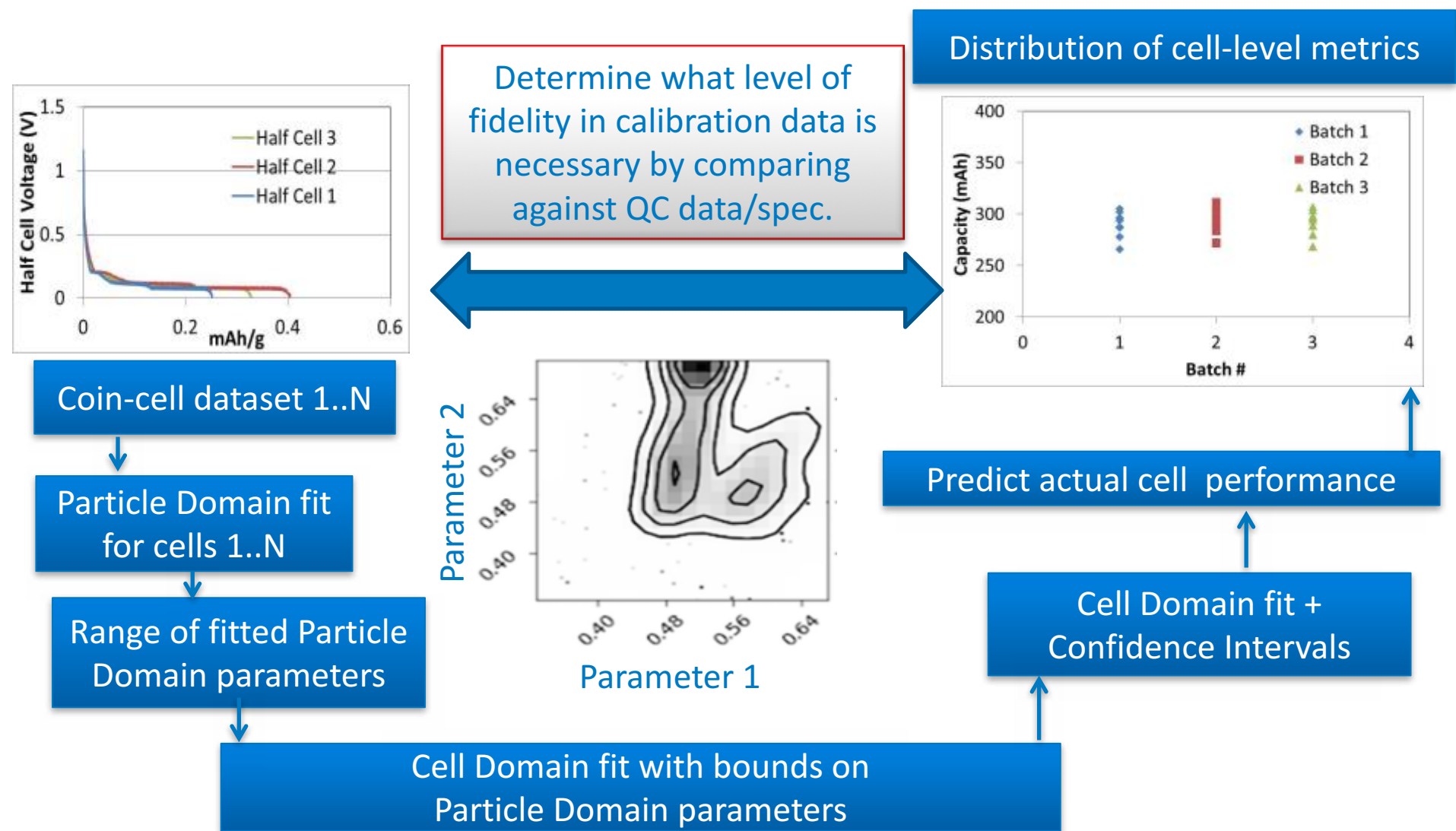CAMP: Cell Analysis, Modeling, and Prototyping

GITT: Model vs. Data

Sample Half-Cell Fits

Sample Full-Cell Validation
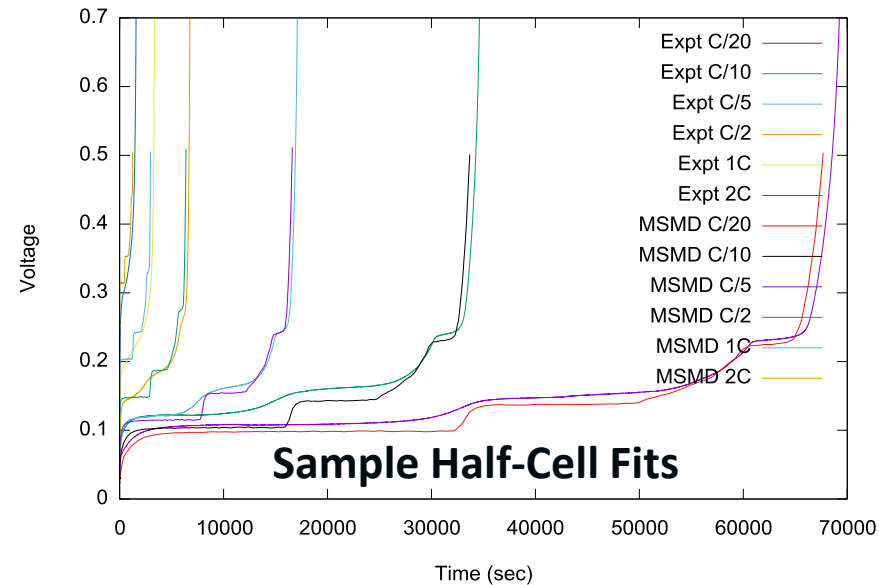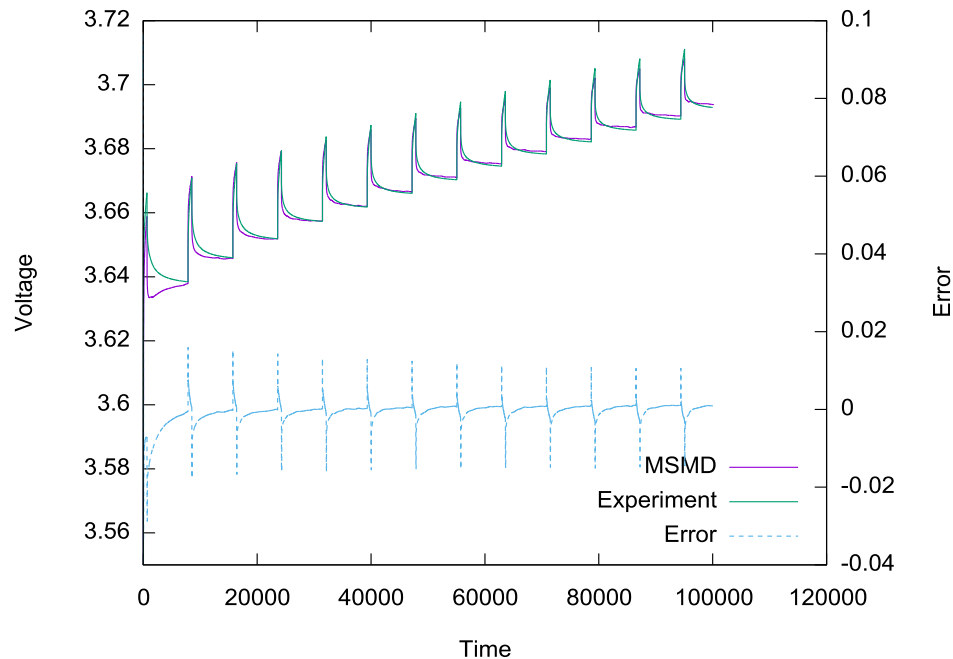
Examples of Parameters and Confidence Intervals

Parameter	Anode	Cathode
C_{Li}^{max} (mol/m³)	2.9511e+04 $\pm$ 2.5377e+02	4.9050e+04 $\pm$ 7.0677e+01
D_s^{ref} (m²/s)	3.015e-15 $\pm$ 2.469e-15	4.393e-15 $\pm$ 2.5634e-17

Automated procedure calibrates models with data from cyclers to a max. relative error < 5%

Application: Analysis of Material/Data Quality

Determine what level of fidelity in calibration data is necessary by comparing against QC data/spec.

Distribution of cell-level metrics

Coin-cell dataset 1..N

Particle Domain fit for cells 1..N

Range of fitted Particle Domain parameters

Predict actual cell performance

Cell Domain fit + Confidence Intervals

Cell Domain fit with bounds on Particle Domain parameters

Closing the loop between lab-scale calibration data and production cell specs. will reduce development costs by directing improvements to processes that impact on cell quality the most.
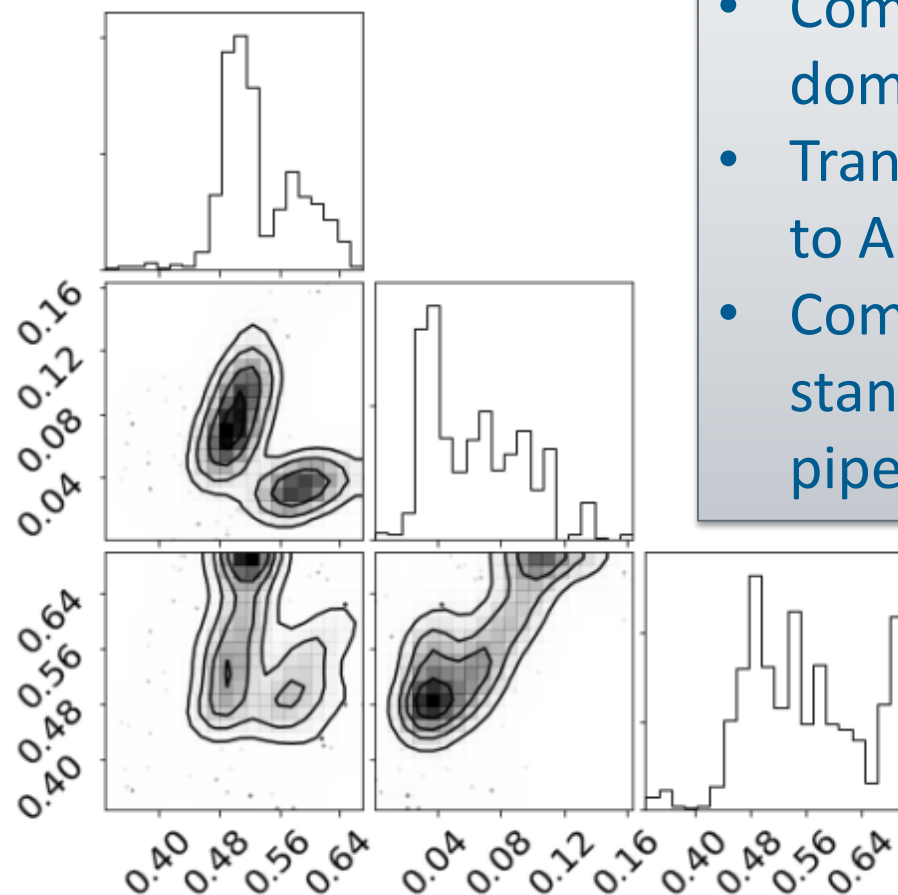
Multi-level Bayesian calibration

Computational Efficiency:
- Complete validation for the cell-domain in FY17
- Transfer GH-MSMD capabilities as UDF to ANSYS Models
- Complete generalization and standardization of the automation pipeline for model identification

Parameter Identification:
- Multiple data sets: what quality of data is needed to induce a given confidence level in the parameters?

Any proposed future work is subject to change based on funding levels.

Mechanical Modeling Approach

<u>Objective</u>: Predict battery behavior during a crash event to optimize safety and weight reduction

Displacement under Crush
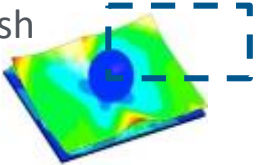
Step 2: Explicit Simulations Parameterize Material Response

Predicts cell temperatures to $\pm 10^{\circ}C$

Step 4: Scale to Module-Level
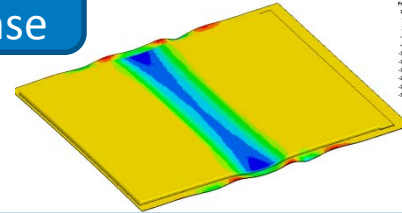
Step 3: Simulate Cell-Level Response for Multiple Cases

Current density under short-circuit

Photo Credits: Jim Marcecki, Ford

Step 1: Start with Component and Cell-Level Test Results as Input
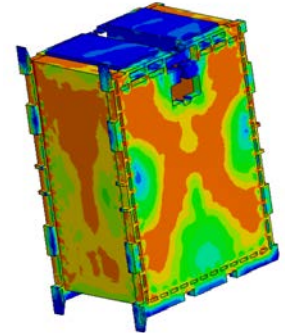
Step 5: Validate against Experimental Data

Goal: Identify localized failure modes and onset loads to within 30 MPa

Sample Input:

- Stress-strain curves for cell components (separator, current collector, etc.)
- Failure strengths for particles
- Mechanical data for cell packaging
- Temperature vs. C-rate for cell
- Abuse reaction data from calorimetry for specific chemistries

Sample Output:

- Current distribution among the different cells within the module
- Localized heat generation rates far away from damage zone
- Stress distribution across multiple parts of the battery module
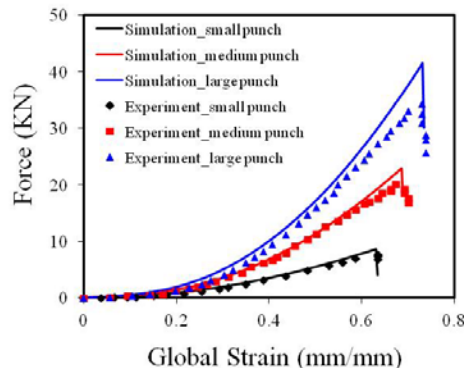
Constitutive Model Development

Step 1. Develop physics-based component models

$$\sigma_{ij,j} + \rho f_i = \rho u_{i,tt}$$

$$\sigma_{ij} = C_{ijkl} \gamma_{kl}$$

$$E = \begin{cases} E_{max} e^{\beta(\varepsilon - \varepsilon_p)} & \varepsilon < \varepsilon_p \\ E_{max} & \varepsilon \geq \varepsilon_p \end{cases}$$
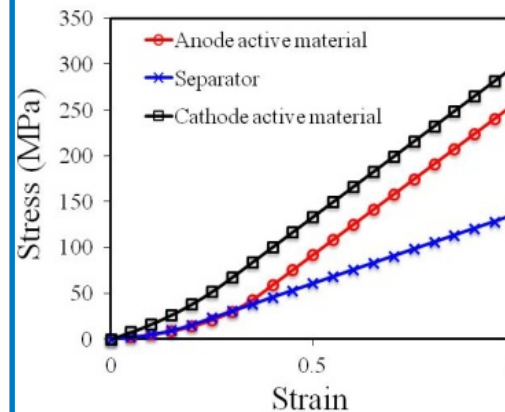
Step 2. Obtain model parameters

Approach a:
Calibrates parameters out of component-level stress-strain data

Approach b:
Phenomenological models for material properties

Legend:
- Anode active material
- Separator
- Cathode active material

Axes: Stress (MPa) vs Strain

Legend:
- Yld-2000 2d
- Von Mises

Axes: σ_{TD}/σ_{eq} [-] vs σ_{MD}/σ_{eq} [-]
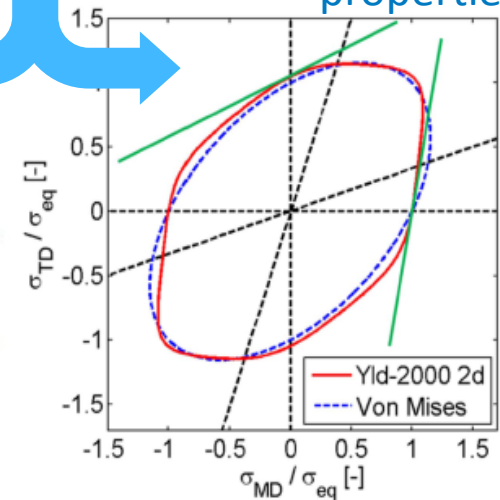
$$\bar{\sigma}(\sigma) = \bar{\sigma}\left(\sigma_{xx}, \sigma_{yy}, \sigma_{xy}\right) = \frac{1}{2^{1/a}}\left(|S_I' - S_{II}'|^a + |2 \cdot S_I'' + S_{II}''|^a + |S_I'' + 2 \cdot S_{II}''|^a\right)^{1/a}$$

Step 3. Validate against independent dataset

Legend:
- Simulation_small punch
- Simulation_medium punch
- Simulation_large punch
- Experiment_small punch
- Experiment_medium punch
- Experiment_large punch

Axes: Force (KN) vs Global Strain (mm/mm)

Cell-level data vs. Model

Macro-scale 3D homogenized mechanical-thermal model

Meso-scale quasi-3D mechanical-thermal model

Pseudo 2D electrochemical-thermal model

Element of the macro-scale model

$d\overline{\varepsilon}, dt, t, T$

$\overline{\sigma}, \sigma_i, \overline{\varepsilon}, \varepsilon_i, T'$

Anode

Separator

Cathode

$R_{short}, Ks_i, dt', t', T$

T'

$$\sigma_{ij} = C_{ijkl}\varepsilon_{kl}$$

Approach for Coupling Methodology:

- Retain fidelity of damage models at the component level (e.g., separate failure criteria for separator, current collector, etc.)
- Solve for potential and temperature as additional degrees of freedom at the component scale
- Simulate multi-cell effects using a micro-mechanical homogenization scheme

$$\begin{Bmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{12} \\ \sigma_{33} \\ \sigma_{23} \\ \sigma_{13} \end{Bmatrix} = \begin{bmatrix} C_{11} & C_{12} & C_{16} & C_{13} & C_{14} & C_{15} \\ C_{12} & C_{22} & C_{26} & C_{23} & C_{24} & C_{25} \\ C_{16} & C_{26} & C_{66} & C_{36} & C_{46} & C_{56} \\ C_{13} & C_{23} & C_{36} & C_{33} & C_{34} & C_{35} \\ C_{14} & C_{24} & C_{46} & C_{34} & C_{44} & C_{45} \\ C_{15} & C_{25} & C_{56} & C_{65} & C_{45} & C_{55} \end{bmatrix} \begin{Bmatrix} \varepsilon_{11} \\ \varepsilon_{22} \\ \varepsilon_{12} \\ \varepsilon_{33} \\ \varepsilon_{23} \\ \varepsilon_{13} \end{Bmatrix}$$

$$\begin{Bmatrix} \sigma_= \\ \sigma_\perp \end{Bmatrix} = \begin{bmatrix} C_= & C_\times^T \\ C_\times & C_\perp \end{bmatrix} \begin{Bmatrix} \varepsilon_= \\ \varepsilon_\perp \end{Bmatrix}$$

Zhang, Chao, et al. *IJES* 2016

40-Ah PHEV cells (NMC-LMO/Gr) were cut open to characterize components

Compression Tests

Before   After

Tension Tests
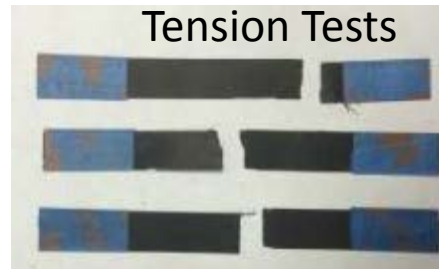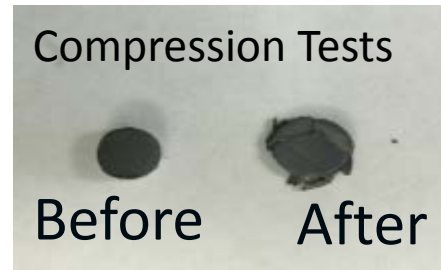
- Failure strains for each cell component under compression and tension were measured
- Properties for the active material were regressed based on composite structure response
- Constitutive model equations represent composite response reasonably well (errors in ultimate stress values < 10%)

Wu, et al. *JES* 2017 (Under Review)
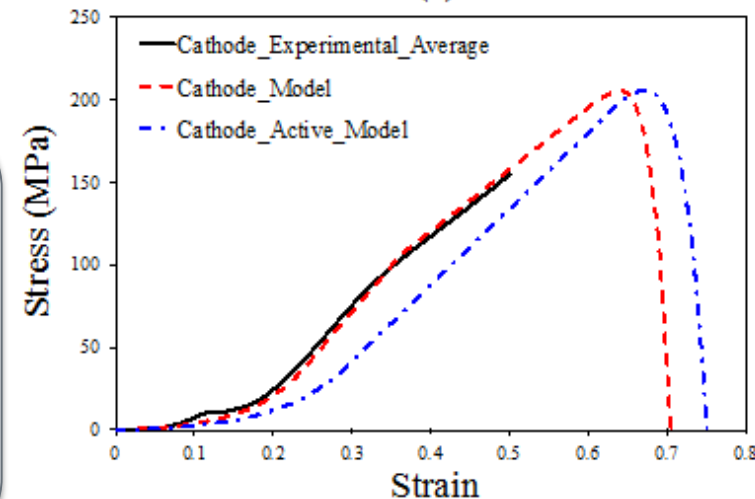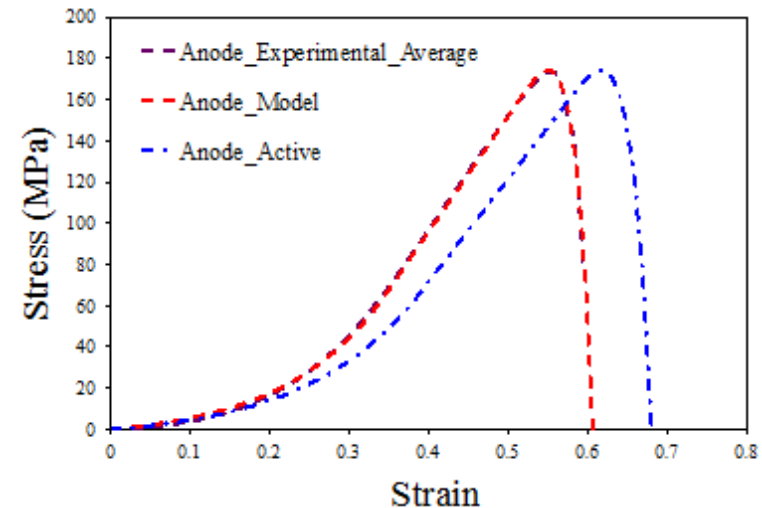
Constitutive response of electrode composites: Model vs. Data

Mechanism of Failure Initiation following a Crush

Side facing the indenter

1st layer | 4th layer | 7th layer

0.72 mm

0.60 mm

Circular tensile crack initiates at strain level 0.36

1 mm

Compression failure

Z-Displacement

-6.939e-17
-5.000e-02
-1.000e-01
-1.500e-01
-2.000e-01
-2.500e-01
-3.000e-01
-3.500e-01
-4.000e-01
-4.500e-01
-5.000e-01

Copper foil Layer 1
Failure of Copper Foil

Anode Layer 4

Cathode Layer 6
Cathode-Anode Short

Cell-level crush tests used to have a "pass" or "fail"

Shear failure of active material layers within a battery

C. Zhang, et al., J. Power Sources, Accepted (Mar. 2017)

Outcome:

- Comprehensive understanding of failure thresholds and propagation mechanism for each component within the cell
- Better explanation of test data results and recommendations for test-methods
- Light-weighting/right-sizing of cells without compromising safety

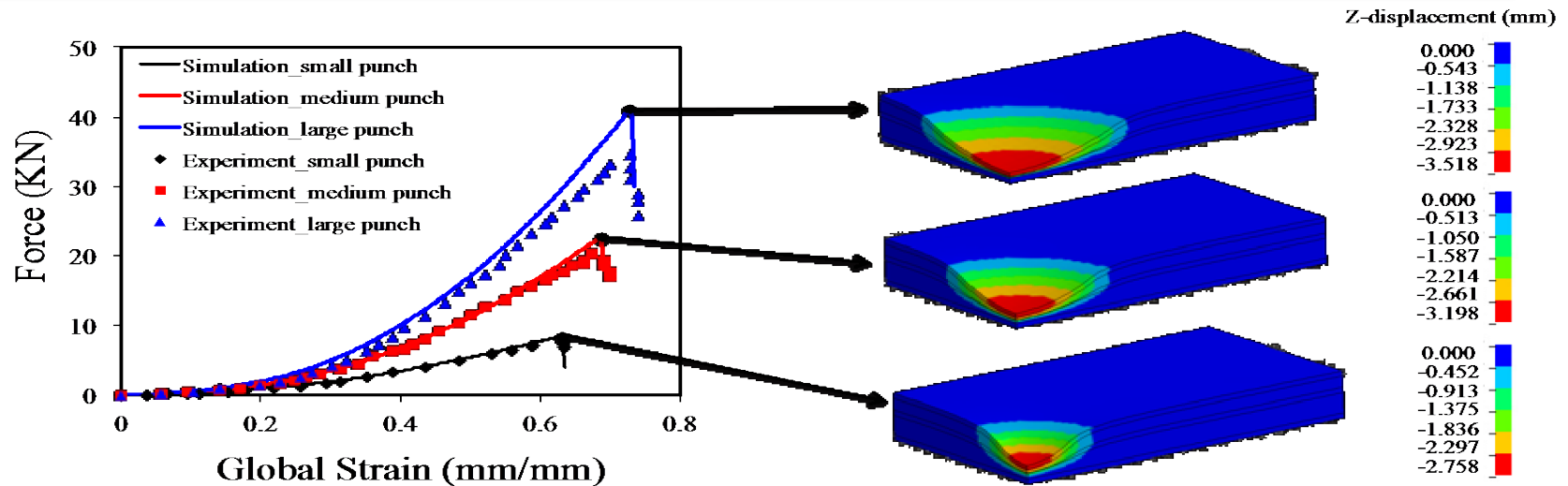Copper foil fails before separator ruptures

Sahraei et al. Journal of Power Sources, 2014

T$_{max}$= 1458°C

T$_{max}$= 224°C

Anode-to-Aluminum Short
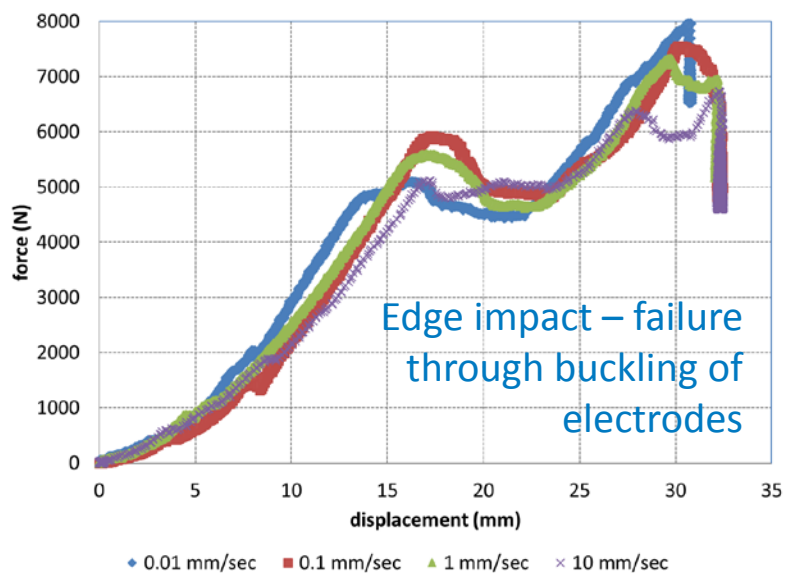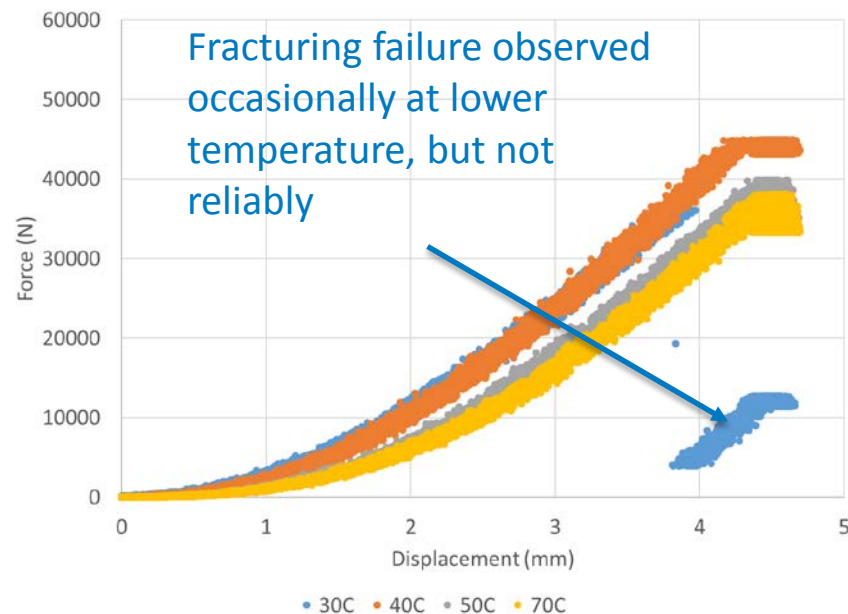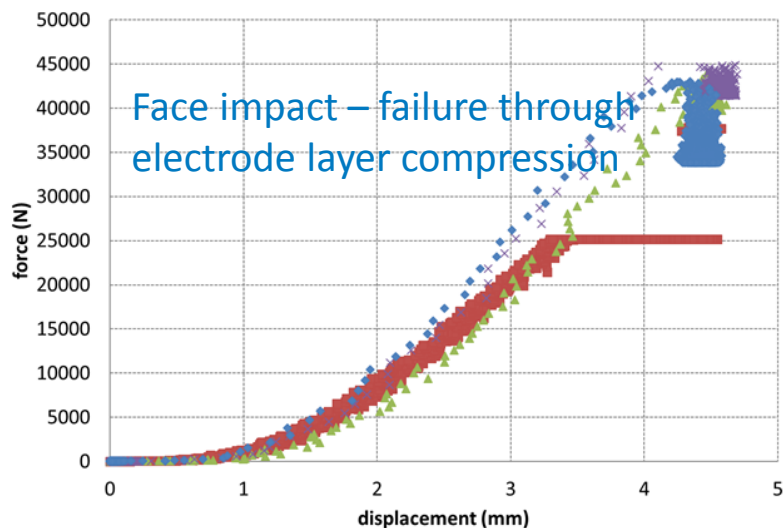
Anode-to-Cathode Short
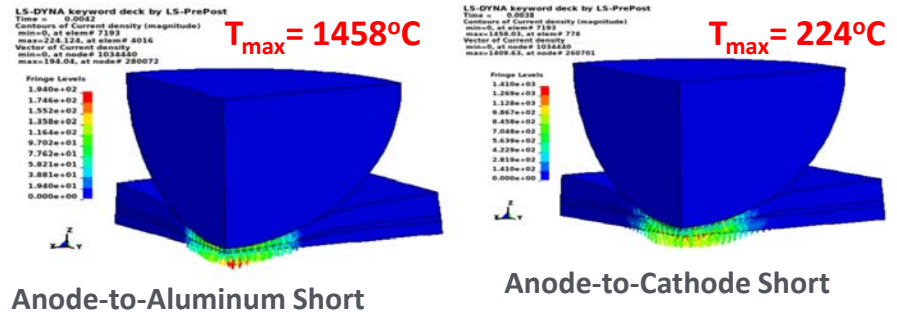
Cell Thermal Response under various types of short-circuit

S. Santhanagopalan, Presented at the International Battery Seminar & Exhibit, 2017.

Models adequately capture mechanical and thermal response under different test conditions.

Single-Cell Test Results from SNL

Face impact – failure through electrode layer compression

0.01 mm/sec   0.1 mm/sec   1 mm/sec   10 mm/sec

Fracturing failure observed occasionally at lower temperature, but not reliably

30C   40C   50C   70C

Edge impact – failure through buckling of electrodes

0.01 mm/sec   0.1 mm/sec   1 mm/sec   10 mm/sec

- Detailed characterization for different orientations, loading rate, temperature
- Some reduced resistance to compression was observed, particularly above 50°C
- Minimal changes with strain rates for range of conditions (0.1–10 mm/s) studied
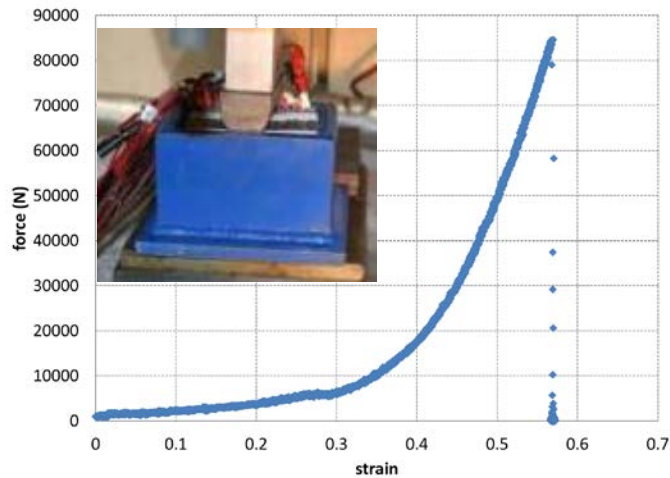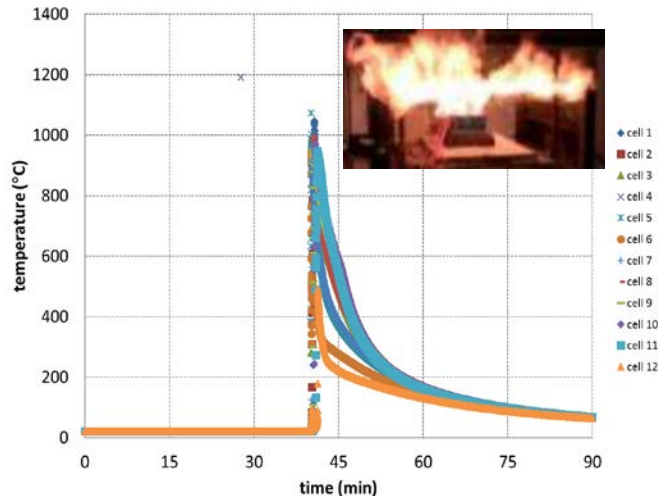- Numerical validation of models initiated

Photo Credits: Joshua Lamb, SNL
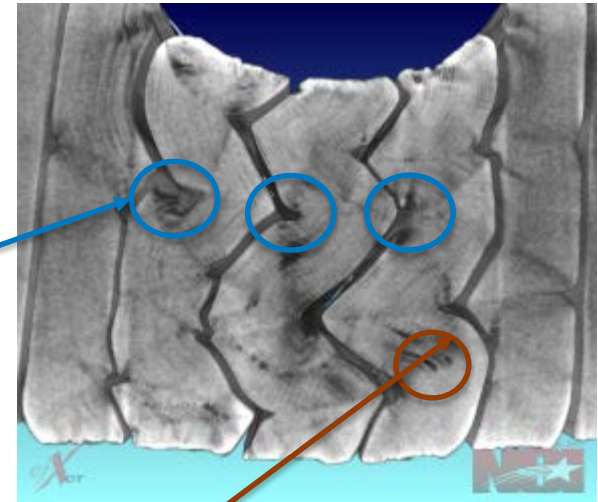
Bar crush of a 12-cell string
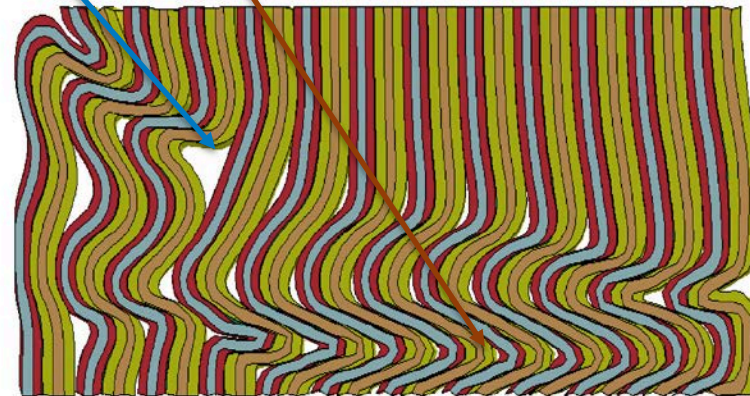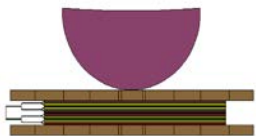

Data

Compression from buckling

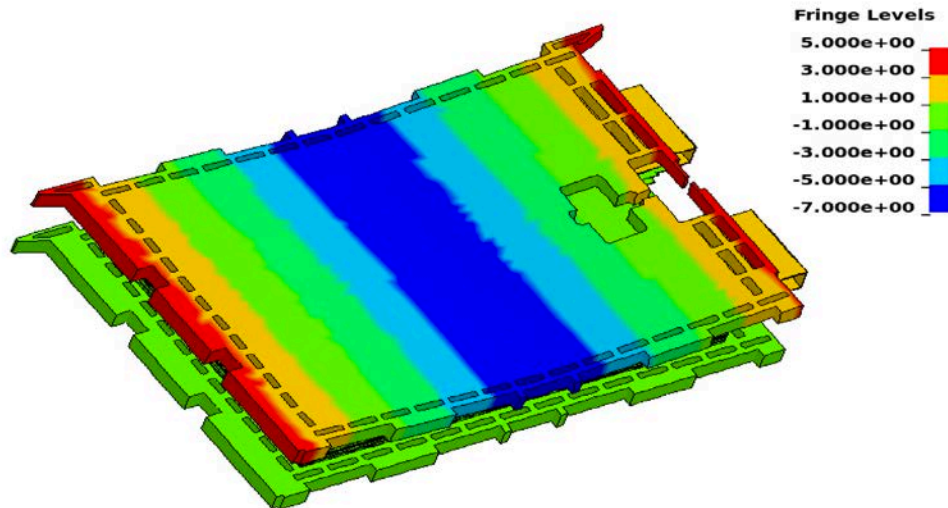**Separation of electrode layers**

Model

Models capture qualitative features; numerical comparison of failure strains underway.
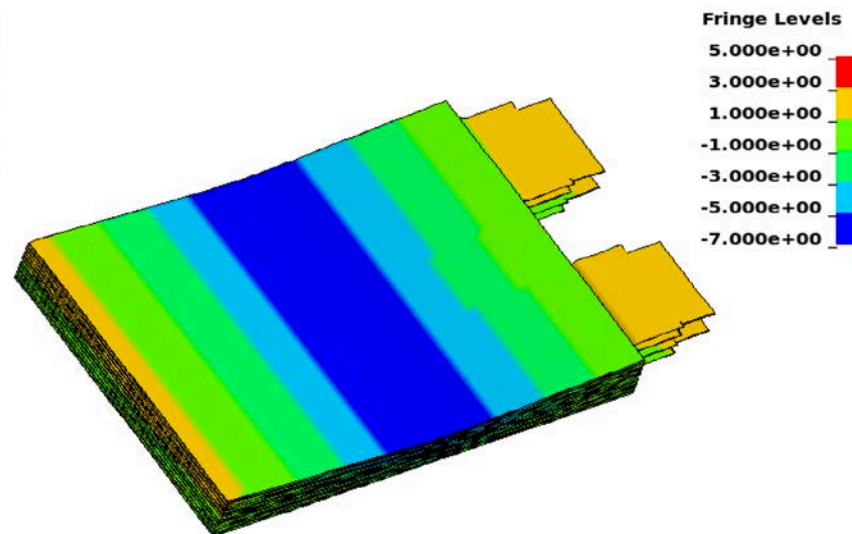
S. Santhanagopalan, Presented at the
International Battery Seminar & Exhibit, 2017.

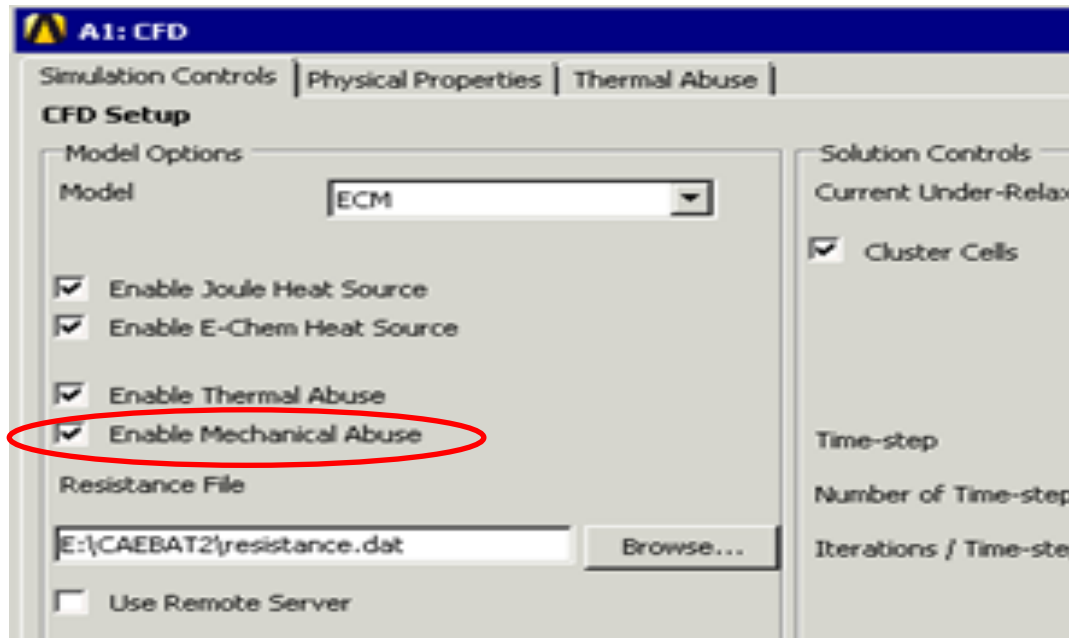**Deformation of packaging material**

Deformation of the cells

Models show that:
- The packaging can prevent deformation of the cells by as much as 50% under these crush test conditions.
- There is a significant scope to lightweight the pack, even after the safety threshold is met.

- Publish procedure for building constitutive relationships for the mechanical models for battery electrodes (2017)
- Complete validation against cell-level and multi-cell data from SNL (2018)

Mechanical Plug-in for ANSYS

- Full cell numerical studies comparing sequential and simultaneous coupling approaches (2018)
- Develop plug-ins to link with other CAEBAT models in ANSYS/ LS-DYNA user-defined models (2018)

Any proposed future work is subject to change based on funding levels.

Response to Previous Year Reviewers' Comments

- <u>Comment</u>: The modeling approach involves building a front-end to commercial solvers (e.g., Fluent), which is very useful for the industry. However, will the tools be accessible to academic researchers without access to these large commercial codes?

  <u>Response</u>: In response to this reviewer's comment, we put together a standalone executable version of the models, which is available to academia and industry alike, for use without requiring licenses to commercial CFD packages. This version is arguably limited in capabilities, but allows end-users to perform quality 3D battery simulations with reasonable computational resources.
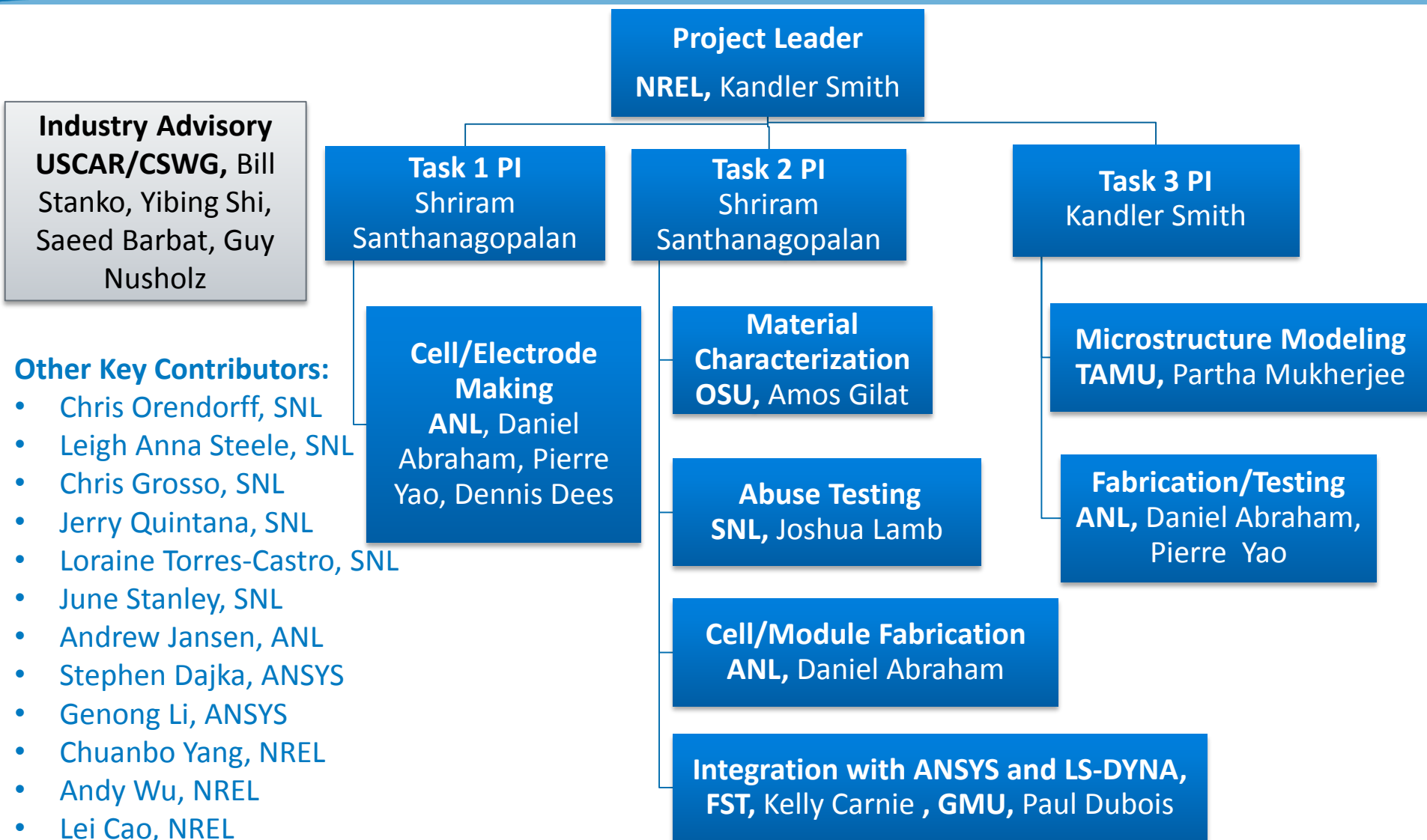
- <u>Comment</u>: Can this effort leverage the project lead by the GM/ANSYS team, co-funded by the Army and the DOE to increase the pack level combined mechanical/electrochemical/ thermal modeling efficiency?
  <u>Response</u>: Yes. The team is working separately with ANSYS to implement the GH-MSMD method into Fluent to achieve similar speed up of models in commercial software.

- <u>Comment:</u> The reviewer cautioned that the community may start making incompatible predictions using different tools put forward by the ORNL, NREL and Ford teams, which could lead to confusion and slow progress.
  <u>Response:</u> The three teams have similar, but complimentary set of goals. We hold quarterly review meetings and host two joint workgroups between the two lab-teams to eliminate minimize overlap, similar to those raised by this reviewer. We also hold monthly updates with the Crash Safety Work Group that includes participants from Ford, GM and FCA where we open the floor for feedback and review.

- <u>Comment:</u> Mechanical failure is a statistical, not deterministic process in which the presence and intensity of local inhomogeneities may control failure rate.
  <u>Response:</u> We account for this artifact by building safety maps to assess room for error in the event of a mechanical failure. The deterministic models are used to build the safety maps by conducting a constraint function sweep across the parameter space to identify robustness of a given cell design.

Collaborators and Partners

Project Leader
NREL, Kandler Smith

Industry Advisory
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Shriram Santhanagopalan

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Task 3 PI
Kandler Smith

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Material Characterization
OSU, Amos Gilat

Abuse Testing
SNL, Joshua Lamb

Cell/Module Fabrication
ANL, Daniel Abraham

Integration with ANSYS and LS-DYNA,
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Microstructure Modeling
TAMU, Partha Mukherjee

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- Andrew Jansen, ANL
- Stephen Dajka, ANSYS
- Genong Li, ANSYS
- Chuanbo Yang, NREL
- Andy Wu, NREL
- Lei Cao, NREL

Summary

- **Task 1**. The GH-MSMD model that provides 100 – 1000x computational speed-up than the MSMD models for battery electrochemical/thermal simulations, is now available for licensing and can run pack simulations on a laptop
  - Publication: G-H. Kim et al., *J. Electrochem. Soc.,* A1076-1088 (2017).
  - Speed enables direct full model use in parameter identification: an automated pipeline to calibrate model from battery-cycler data is under development.

- **Task 2**. Simultaneously coupled mechanical-electrochemical-thermal model for mechanical abuse simulation
  - Multi-scale model can include multiple failure criteria for each component in a module- or pack-level simulation.
  - Initial set of comparisons against test data at the component level shows good promise for homogenization approach.
  - Comprehensive model validation is ongoing in partnership with SNL.
  - Effort to streamline interfacing with off-the-shelf software tools (ANSYS/LS-DYNA) is underway.
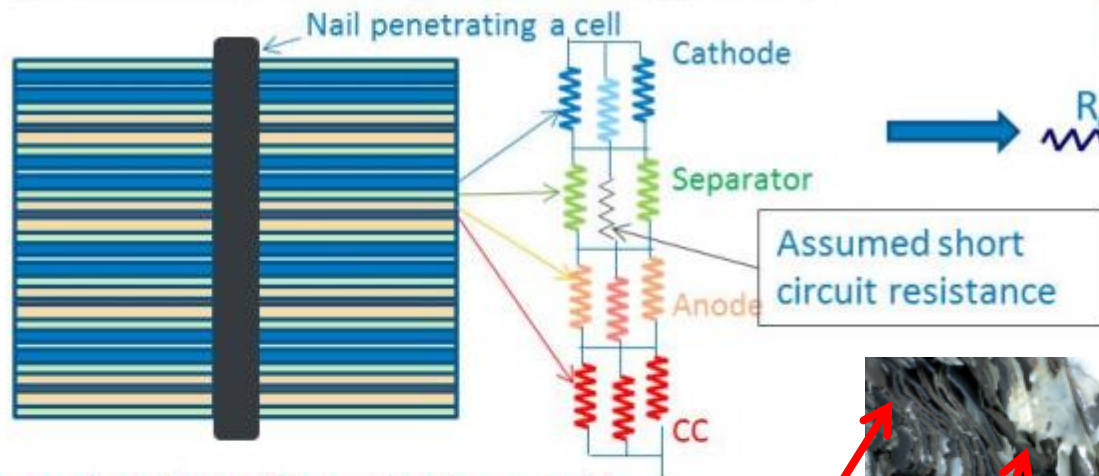
Acknowledgements

- We appreciate support and funding provided by Vehicle Technologies Office at the U.S. Department of Energy

    - Brian Cunningham
    - David Howell
    - Samuel Gillard
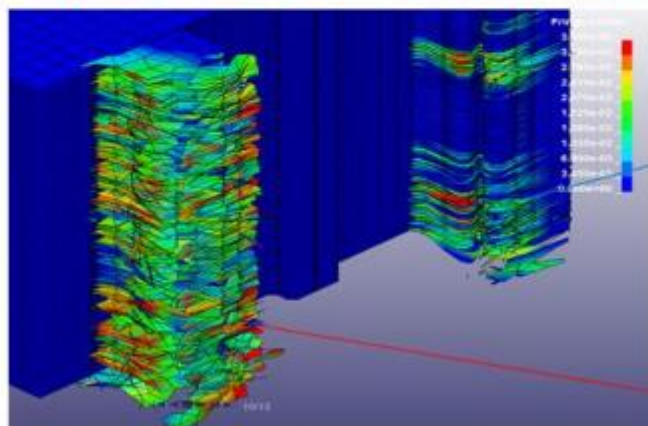
Technical Back-Up Slides
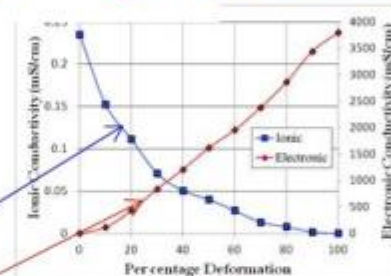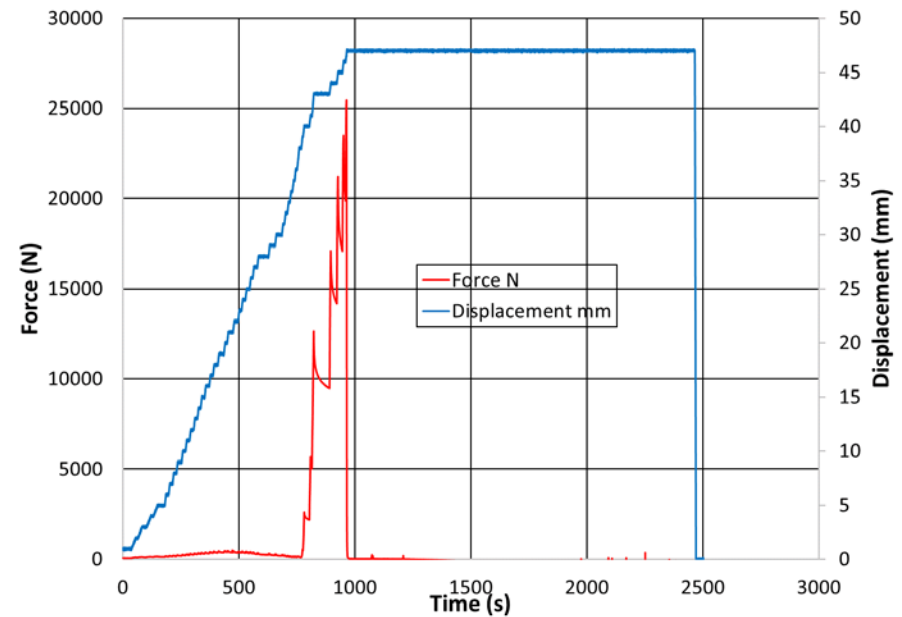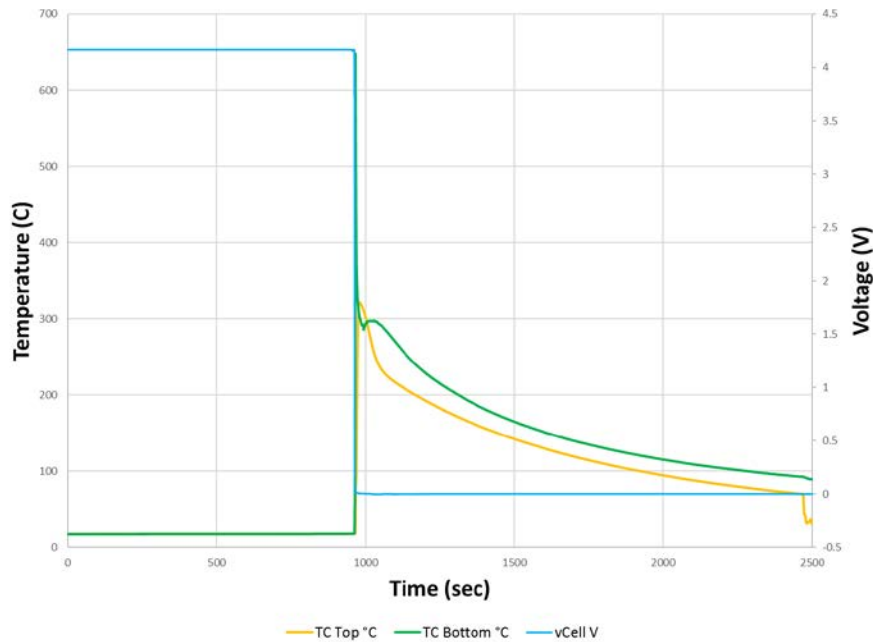
Full charge test

- Initial test conducted in 1-mm steps to determine point of failure
- No failure observed during bend of cell
- Failure achieved through cell compression at end of test
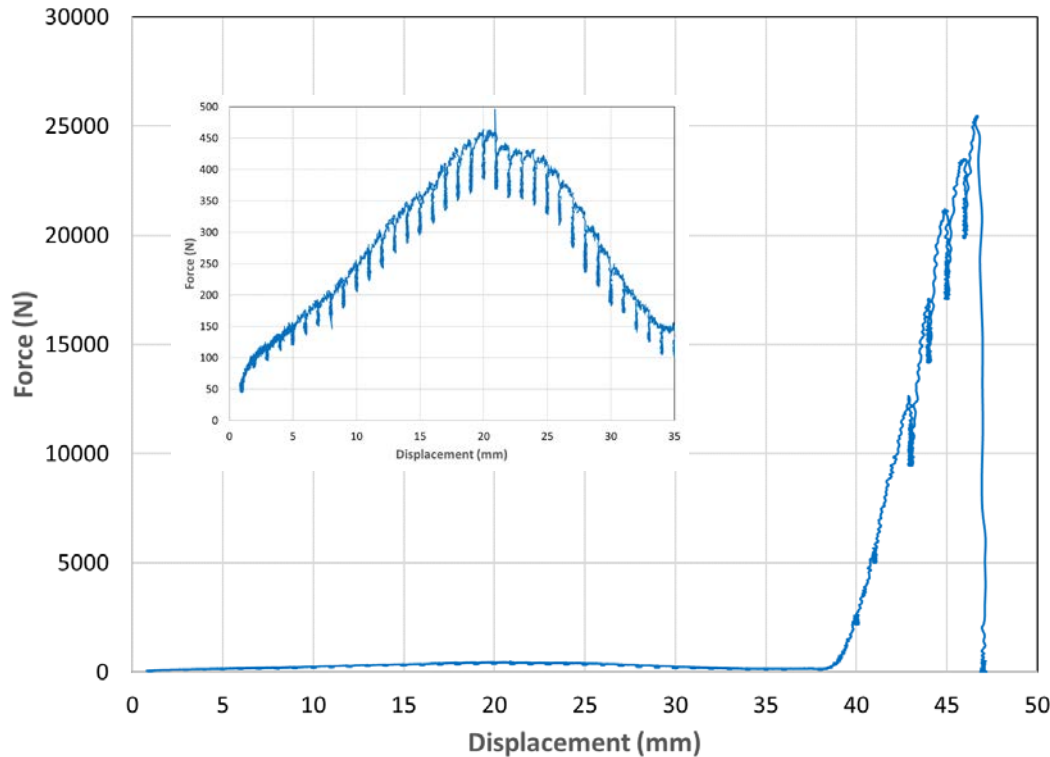
- Bend portion of test shows a yield of ~450 N.
- Cell failure required a compressive force of ~25 kN.
- "Pre-load" portion of bend observed where initial compression is applied to cell before bending occurs.
- After yield of cells to bend the cell is put into compression.

Drop Tower – Impact Tester

Specifications:

- Overall height: 14 feet (4.3 m)
- Drop height: up to 10 feet (3.1 m)
- Drop weight: 50 to 500+ pounds (22.7 – 226.8 kg)
- Max impact velocity ~ 25.4 ft/s (7.74 m/s)
- Impact force (assuming a 6" stopping distance): 10,000 lbs-f (44,482 N)
- Remote operation
- Data collection:
  - Displacement
  - Impactor velocity
  - Force at impact
  - Temperature
  - Voltage

Figure Credit: Joshua Lamb, SNL

Drop Tower – Impact Tester

Current Status:

- ✓ CAD model – complete
- ✓ Drawing package – complete
- ✓ Hardware bill of materials (BOM) – complete
- ✓ Controls box design – complete
- ✓ Controls BOM – complete
- ✓ BATLab personnel to order all controls hardware – near complete
- ○ Build request, including drawing package – submitted to contractor
- • BATLab personnel to order all hardware for build – waiting on contractor readiness
- • BATLab personnel to complete final assembly of drop tower – waiting on completion of contractor build

Figure Credit: Joshua Lamb, SNL